



Attorney's Docket No. 1030681-000521

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re Patent Application of) MAIL STOP AMENDMENT
Won-bong Choi et al.) Group Art Unit: 1795
Application No.: 10/601,872) Examiner: CYNTHIA K LEE
Filed: June 24, 2003) Confirmation No.: 1325
For: CARBON NANOTUBES FOR FUEL)
CELLS, METHOD FOR)
MANUFACTURING THE SAME, AND)
FUEL CELL USING THE SAME)

DECLARATION UNDER 37 C.F.R. § 1.131 BY PATENT COUNSEL

Commissioner for Patents
P.O. Box 1450
Alexandria, VA 22313-1450

Sir:

I hereby declare that:

1. I am employed at Y.P. Lee, Mock & Partners in Seoul, Republic of Korea. I supervise staff, including the staff that wrote the above-captioned application.
2. I was the supervising counsel for the writing of the above-captioned application.
3. Responsibility for drafting the application was transferred to my firm in May of 2002. See Attachments A-C illustrating our receipt in May 2002. The approximate three-month time period in drafting the application (date of filing was July 29, 2002 for the priority document, Korean Patent Application No. 2002-44631) shows reasonable diligence in preparing and filing the patent application as this is approximate to the average time it takes my firm to draft patent applications in the fuel cell technologies for Samsung. There is no indication when the priority was given to any applications in this particular area of fuel cells and the applications were taken up in chronological order. It is my opinion that the persons responsible worked reasonably hard on the particular application in question during the continuous critical period of May 9, 2002 to July 29, 2002.

I hereby declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful, false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the

Buchanan Ingersoll & Rooney PC
Attorneys & Government Relations Professionals

Declaration by Patent Counsel Under 37 C.F.R. § 1.131
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Page 2

United States Code and such willful, false statements may jeopardize the validity of the application or any patent issued thereon.

I hereby declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issued thereon.

Date: 2 April 2008

By: Deymijie Zhou
Title: Patent attorney



CERTIFICATION OF TRANSLATION

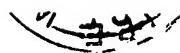
I, Ji Hye Ohn, an employee of Y.P.LEE, MOCK & PARTNERS of Koryo Bldg., 1575-1 Seocho-dong, Seocho-gu, Seoul, Republic of Korea, hereby declare under penalty of perjury that I understand the Korean language and the English language; that I am fully capable of translating from Korean to English and vice versa; and that, to the best of my knowledge and belief, the statement in the English language in the attached translation of Employee Invention Document of Korean Patent Application No. 10-2002-0044631 consisting of 7 pages, have the same meanings as the statements in the Korean language in the original document, a copy of which I have examined.

Signed this 1st day of April 2008

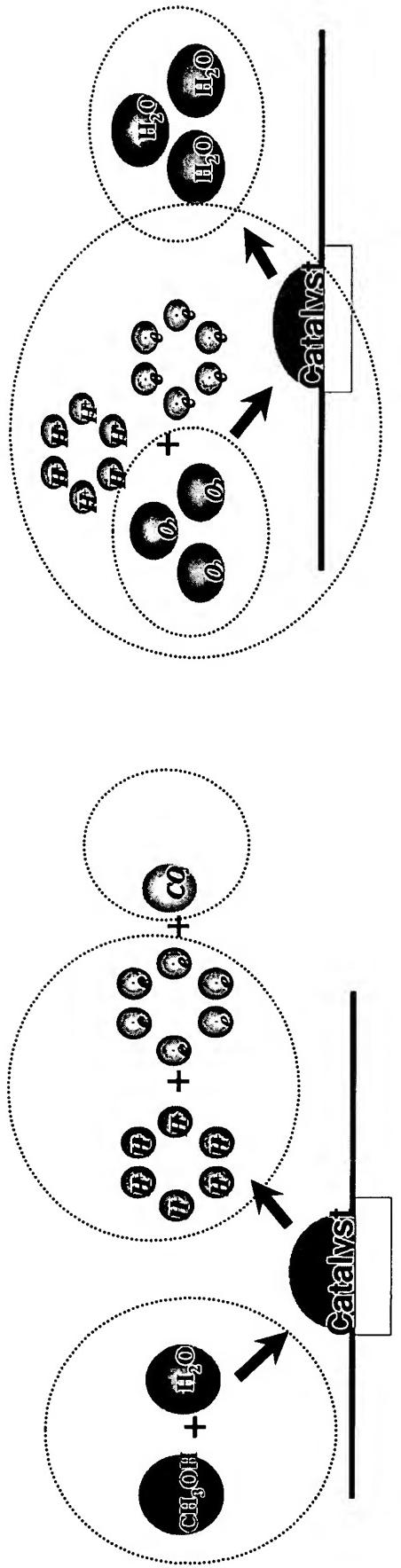
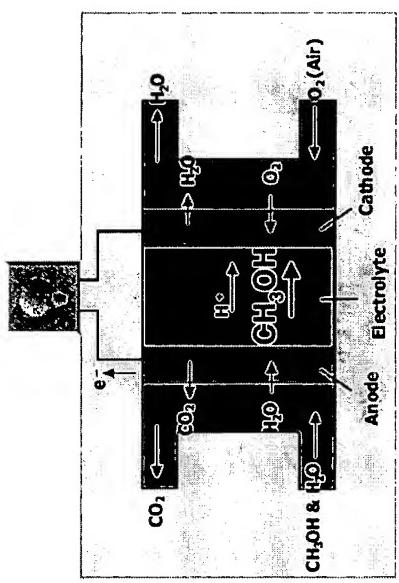
A handwritten signature in black ink, appearing to read "Ji Hye Ohn".

Development of Carbon Nanotube Electrode for Fuel Cell

Inventors : CHOI Won Bong, CHU Jae Uk, PAK Chanho, CHANG Hyuk



Principle Diagram

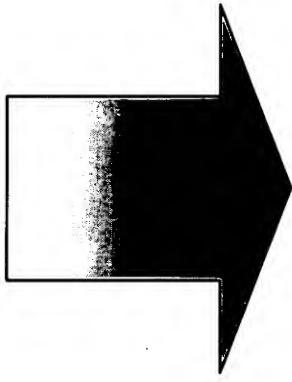


$$E^\circ = 0.015 \text{ V}$$

$$E^\circ = 1.229 \text{ V}$$

Issues

- ☒ Surface area must be large.
- ☒ Catalyst must be stabilized in nano scale and be uniformly dispersed.
- ☒ Catalyst must efficiently react with solution for fuel cells (e.g. methanol).
- ☒ Cost must be inexpensive.



Solutions

1. Catalyst is uniformly dispersed on the inner and outer walls of CNT during CNT growth. (Catalyst nanoparticles are uniformly distributed between carbon atoms)
2. Inner and outer walls of CNT are utilized to maximize the surface area
3. CNT are grown into a branch type in order to maximize the surface area.
4. Catalyst nanoparticles are stably dispersed and is stable against external influences.
5. CNT are grown directly on a carbon film for electrodes, simplifying the process - Prevent cost increase by introducing a simple carbon nanotube growth method.

Synthesis steps of carbon nanotube

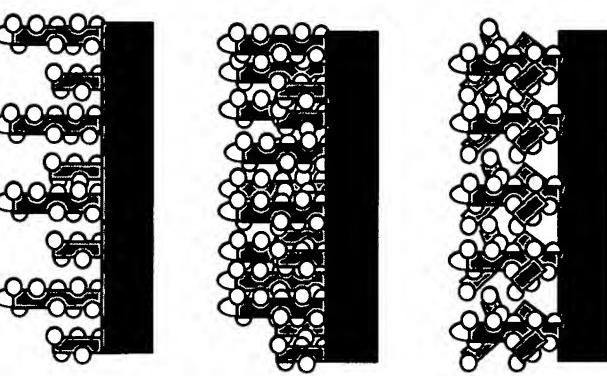
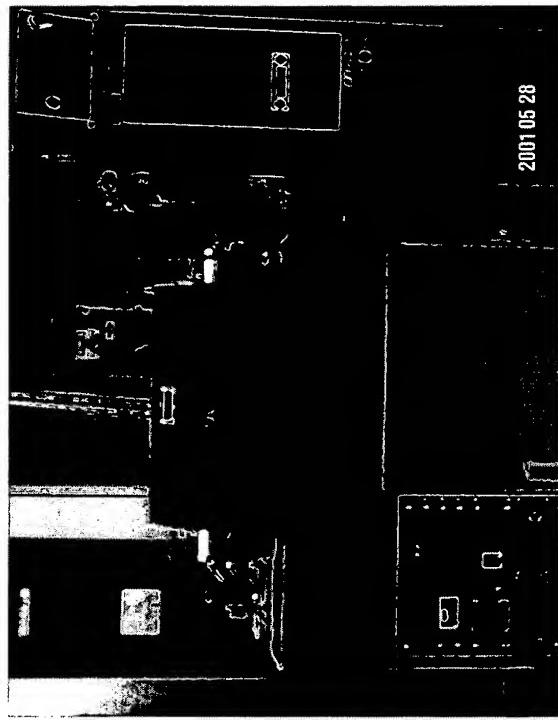
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1. Uniformly dispersing the catalyst

0 0 0 0 0 0
2. Growing CNT while
dispersing the catalyst

0 0 0 0 0 0
3. Controlling the density during
CNT growth

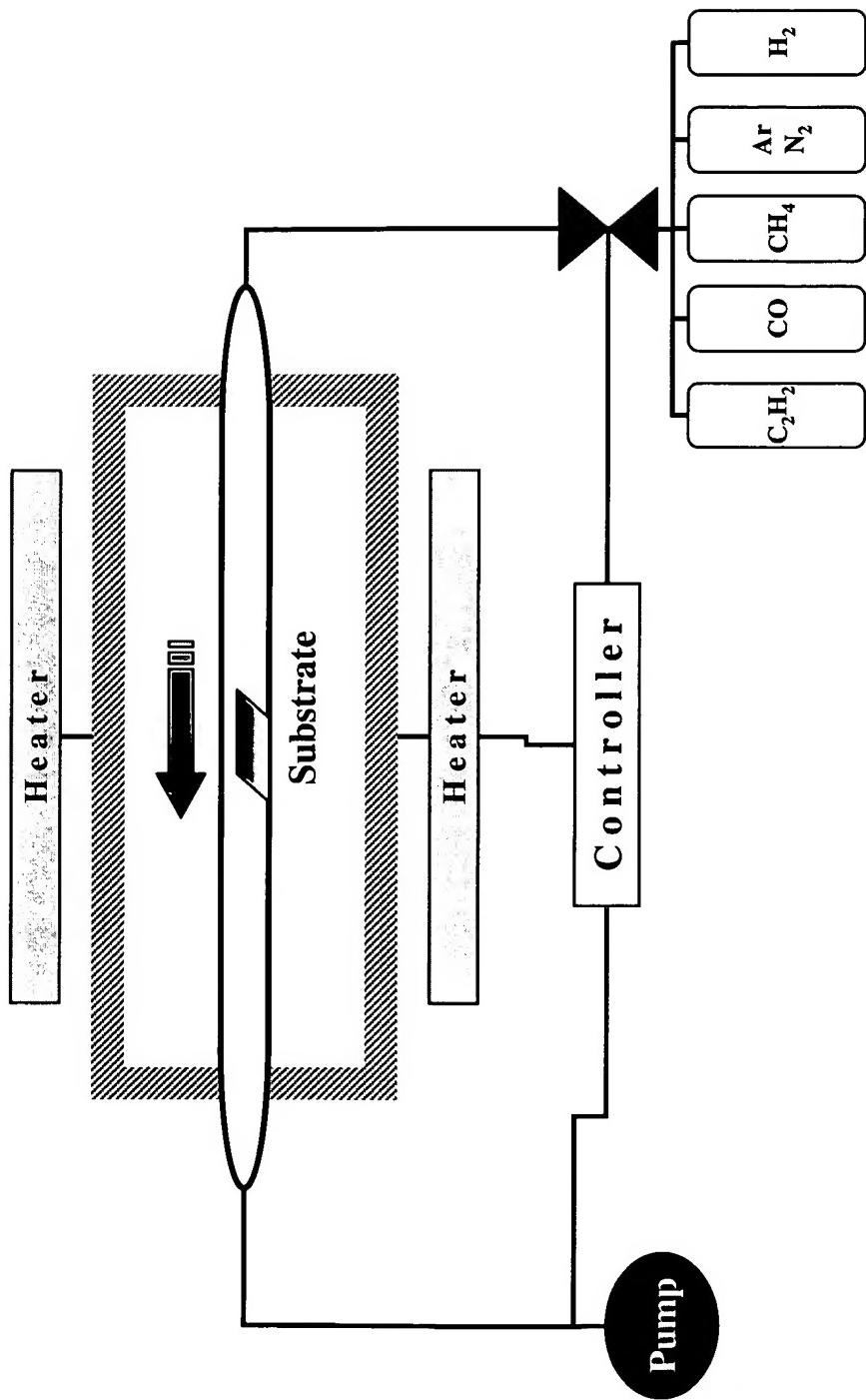
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3-1. Increasing the surface area
by branching out CNT during
growth

Rapid thermal CVD (S.A.I.T)



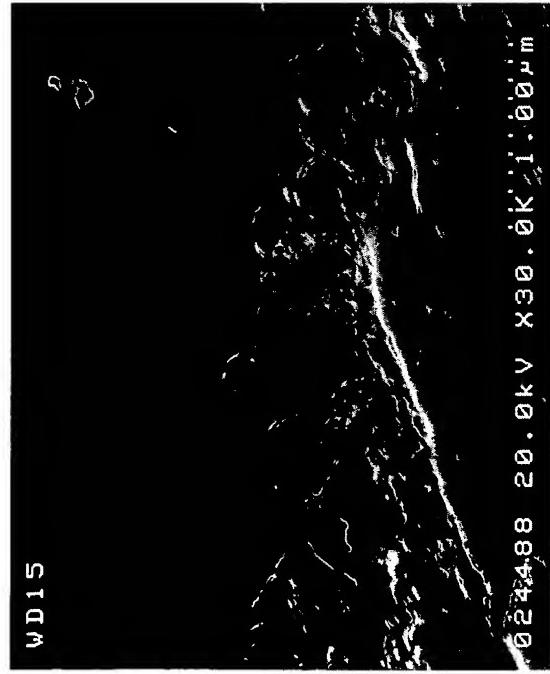
Schematic of carbon nanotube-Synthesis

- Thermal CVD : Reaction temp. 500-900°C, Time 1min - 30min



Features of SAIT Carbon Nanotube Electrode

1. Process is simplified by growing directly on carbon film for electrode, because of omission of a process of manufacturing catalyst support and electrode.
2. Catalyst is uniformly dispersed on the inner and outer walls of CNT to maximize the surface area for catalytic reaction.
3. Catalyst nanoparticles are stably dispersed, and unaffected by external influences.
4. CNT shape can be modified during growth (Maximization of surface area).



SEM image of grown CNT

Claims

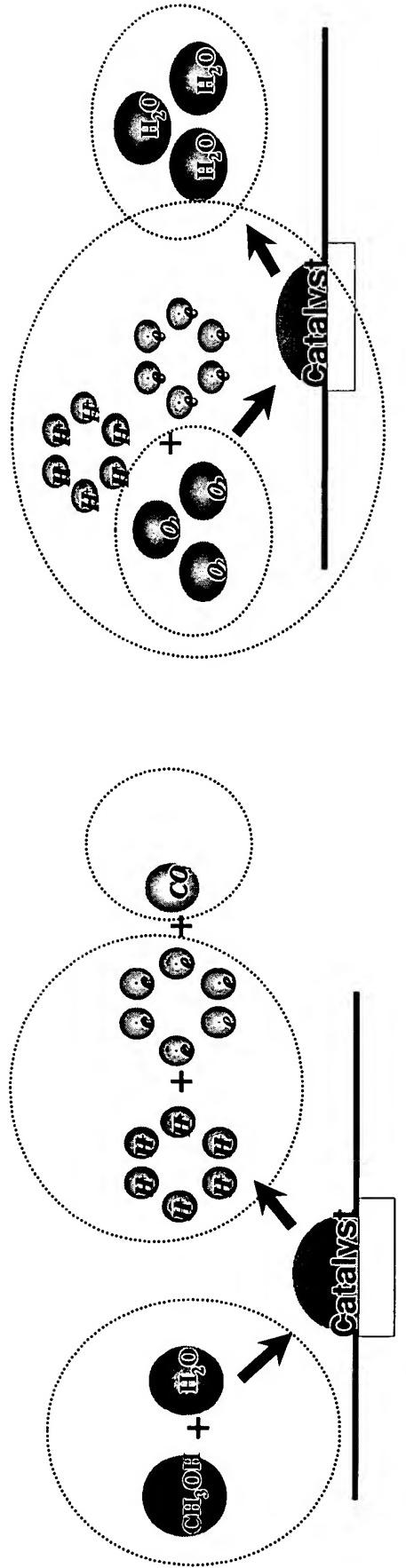
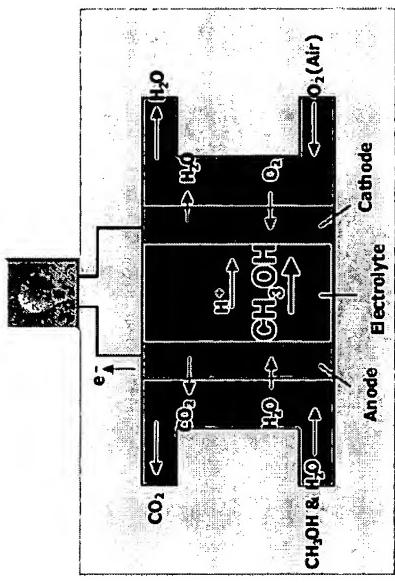
- ☞ A process of growing carbon nanotubes having nano-sized catalyst particles (Pt, Ru, Fe, Co, etc.) uniformly dispersed on the inner and outer walls of the carbon nanotubes
- ☞ A process of directly growing carbon nanotubes with catalyst dispersed on a carbon film for electrodes
- ☞ A concept of using Pt, Ru, Fe, Co etc., or binary, ternary, and quaternary alloys thereof as catalysts
- ☞ A process of pretreatment (by Electrophoresis, Thermal Spray, Sputtering, or CVD etc) which uniformly disperses carbon nanotube-growing catalysts (Pt, Ru, Fe, Co etc)
- ☞ A concept of using the carbon nanotube with the catalysts adhered thereto as the cathode and the anode

Fuel Cell 用 탄소 나노튜브 전극 개발

발명자 : 최원봉, 주제욱, 박찬호, 장혁



운|리|도



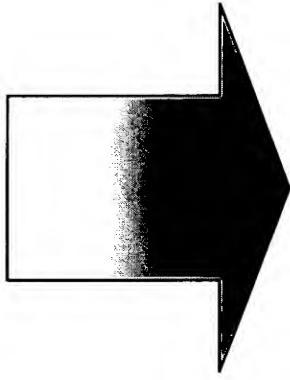
$$E^\circ = 0.015 \text{ V}$$



$$E^\circ = 1.229 \text{ V}$$

Issues

- ☞ 비표면적이 커야 한다.
- ☞ 측매가 나노 사이즈로 안정화 되어 균일하게 분산되어야 한다.
- ☞ 측매가 연료전지용 용액(메탄올 등)과 반응을 효율적으로 하여야 한다.
- ☞ 가격이 비싸지 않아야 한다.



Solutions

1. CNT성장시 측매를 CNT 내.외벽에 균일하게 분산시킴.
(나노입자의 측매가 탄소원자 사이에 균일하게 분포된 구조.)
 2. CNT 내.외벽을 활용 측매반응의 비표면적을 최대화 시킴.
 3. CNT성장시 가지형으로 성장시켜 비표면적을 크게 한다.
 4. 나노입자의 측매가 안정하게 분산되어 외적인 영향에 안정됨.
 5. 전극용 탄소막에 직접 성장시켜 공정 단순화
- 간단한 탄소나노튜브 성장 방법을 도입함으로써 가격상승 억제

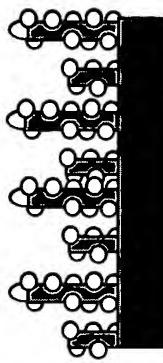
Synthesis steps of carbon nanotube

1. 촉매를 균일하게 분산시키는 공정

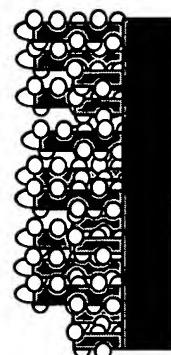


Rapid thermal CVD (S.A.I.T)

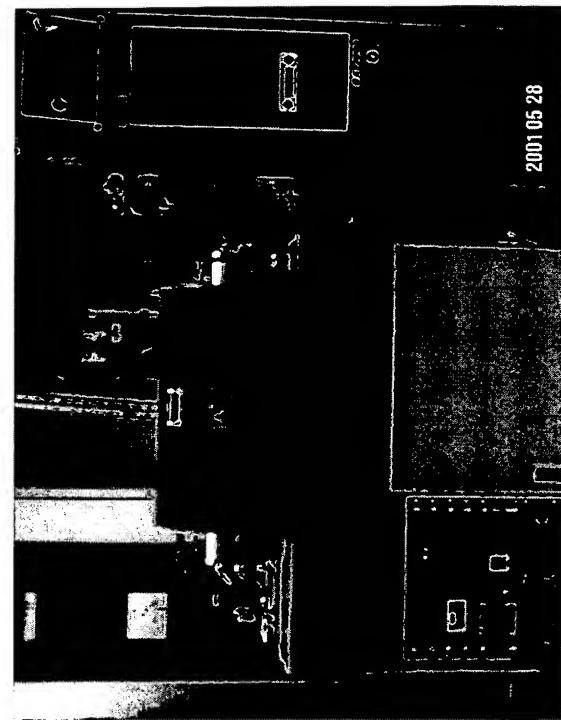
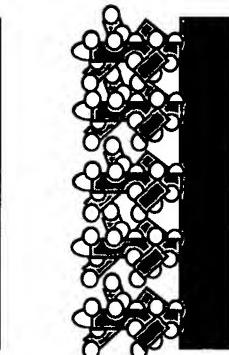
2. 촉매를 분산시키면서
CNT성장시키는 공정



3. CNT성장 시 밀도를 조절하는 공정

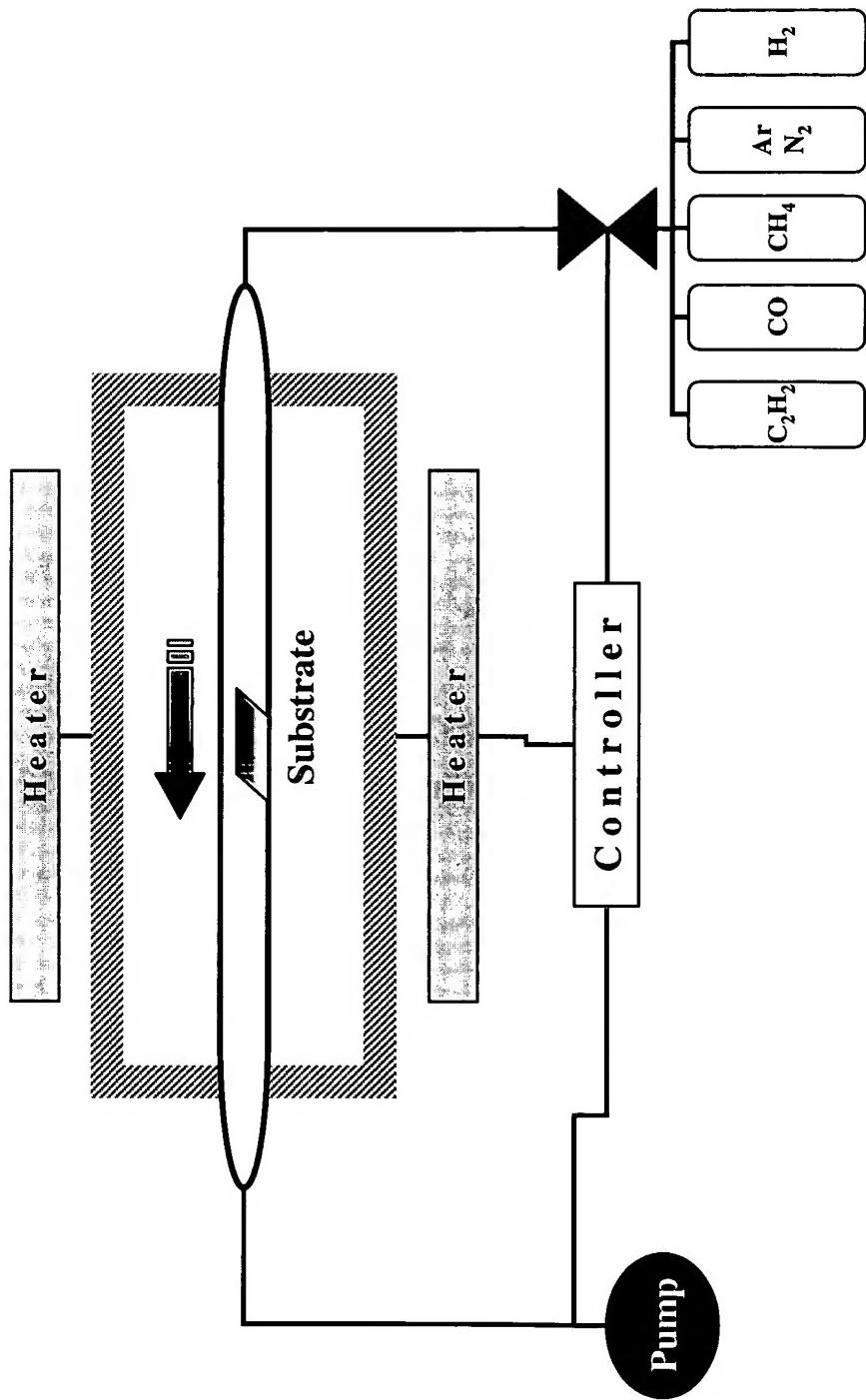


3-1. CNT의 가지를 치면서 성장하여
비표면적을 넓히는 공정



Schematic of carbon nanotube-Synthesis

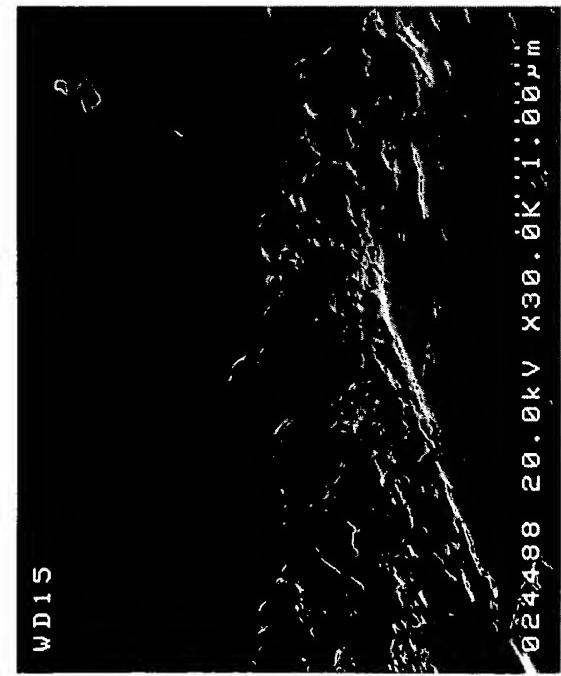
- Thermal CVD : 반응온도 500-900°C, 시간 1min - 30min



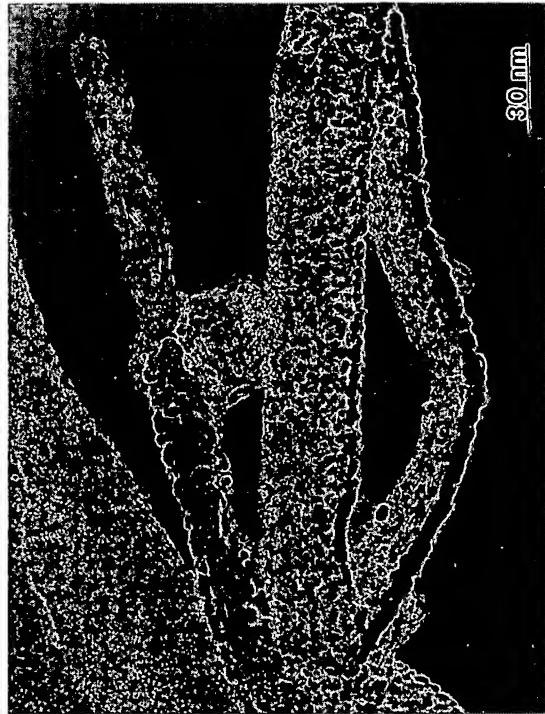
다|오|비

기술원 탄소나노튜브 전극 특징

1. 전극용 탄소막에 적절 성장시켜 측매답지, 전극제조 과정 생략으로 공정 단순화.
2. CNT 내부 및 외벽에 측매를 균일 분산시켜 측매반응의 비표면적을 최대화 시킴.
3. 나노입자의 측매가 안정하게 분산되어 외적인 영향에 따른 유동이 없음.
4. CNT 성장시 모양 변형 가능 (표면적 증대 효과).



CNT성장된 SEM 사진



CNT성장된 TEM 사진

장 구 험

- ☞ 탄소 나노튜브의 내부 및 외벽에 나노크기의 촉매입자(Pt, Ru, Fe, Co etc)가 균일하게 분산된 구조를 갖는 탄소 나노튜브를 성장시키는 CNT 성장공정.
- ☞ 전극용 탄소필름에 촉매 분산된 나노튜브를 직접성장 시키는 공정.
- ☞ Pt, Ru, Fe, Co등을 촉매로 사용하며, 이들의 이원계, 삼원계, 사원계 합금을 촉매로 이용하는 개념.
- ☞ 탄소 나노튜브 성장용 촉매(Pt, Ru, Fe, Co등)를 균일하게 분산시키는 전자리 과정(Electrophoresis, Thermal Spray, Sputtering, CVD etc).
- ☞ 상기 촉매가 부착된 탄소 나노튜브를 Fuel cell의 Cathode 및 Anode 전극으로 사용하는 개념.